Observation of an unusual ESR signal in antiferromagnetic Eu_2CuO_4

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We report the observation of an unusual electron spin resonance (ESR) signal in single crystals of Eu_2CuO_4. The signal appears to be associated with a resonance mode of the CuO_2 planes, similar to the midfield and low-field absorptions we have reported previously [Phys. Rev. **B 41**, 1934 (1990)]. However, it is only observed when the projection of the applied dc magnetic field in the CuO_2 plane is within a few degrees of the ⟨110⟩ crystallographic direction. Additionally the sample must be field cooled in the CuO_2 plane, but with a component of the cooling field perpendicular to the ⟨110⟩ ESR observation direction. Both the field for resonance and the linewidth exhibit a $1/\cos\theta$ dependence, where θ is the angle of the applied dc field between the c axis and the ⟨110⟩ observation direction. Additional constraints for observation of the resonance are that the microwave rf magnetic field must have a component in the CuO_2 plane, but perpendicular to the dc field. The signal disappears above $\sim$215 K, which we assume is associated with the antiferromagnetic ordering temperature.

We have previously reported on the unusual and diverse magnetic properties observed in single crystals of the rare-earth copper oxides, R_2CuO_4 with R = Eu or Gd, below the Cu Neel ordering temperature T_N, which occurs at $\sim$270 K.[1] In particular, we have observed two resonant absorptions, which we termed the low-field absorption (LFA) and midfield absorption (MFA). These absorptions exhibited a large out-of-plane anisotropy and were ascribed to resonant modes of the CuO_2 planes. They were not considered typical EPR signals in that the absorptions could be observed for any orientation of the microwave magnetic field h_{rf} to the applied dc magnetic field H_{dc}.

In this paper we report on a newly discovered microwave absorption signal (MAS) in single crystals of Eu_2CuO_4 which were prepared by standard techniques.[1] In contrast to the LFA and MFA, the MAS is not observable if h_{rf} is parallel to H_{dc}. The MAS exhibits the same out-of-plane anisotropy found for the low-field and midfield absorptions, but in addition, it exhibits a strong and unusual anisotropy within the CuO_2 plane. In particular, it is only observed when the dc magnetic field is oriented within a few degrees of the ⟨110⟩ crystallographic direction!

We find that the MAS is sample dependent. Although we have found it in both pure and Ce doped Eu_2CuO_4, the signal is not observed in all of the Eu_2CuO_4 samples which we have investigated. We cannot rule out the possibility that the MAS arises from an impurity within the Eu_2CuO_4 compound, but if so, we are unaware of any prior report of an impurity response as unusual as that presented here.

The signal was detected with a standard EPR superheterodyne spectrometer operating at 9.2 GHz in the field derivative mode. Data was taken at temperatures ranging from 77–300 K. We have also observed the MAS at a frequency of 35 GHz and in Eu_2CuO_4 crystals doped with Gd and/or Ce. These preliminary results, as well as data taken at temperatures below 77 K, will be presented in a future publication.

In Fig. 1 we present spectra taken for a Eu_2CuO_4 single crystal at 77 K with H_{dc} applied along the CuO_2 plane at various angles near the ⟨110⟩ crystallographic direction. In order to observe the MAS, the sample must be cooled below T_N in a large dc magnetic field (discussed below). When H_{dc} is applied within the CuO_2 plane, the MAS

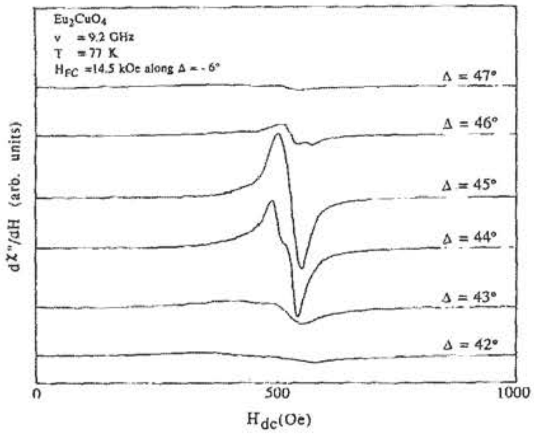

FIG. 1. Spectra for Eu_2CuO_4 taken at various angles, Δ, in the CuO_2 plane, near the ⟨110⟩ direction. The sample was cooled to 77 K in a field of 14.5 kOe applied along $\Delta = -6°$.

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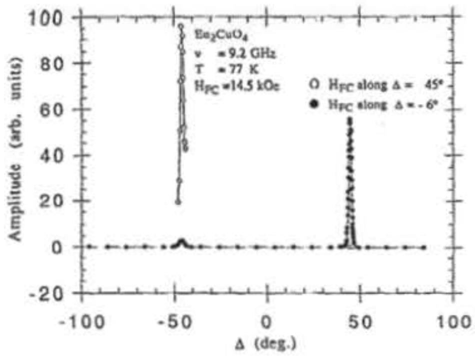

FIG. 2. Microwave absorption signal amplitude as a function of the angle Δ of H_{dc} within the CuO_2 plane. The sample was cooled to 77 K in a field of 14.5 kOe applied along either $\Delta = -6°$ (solid circles) or $\Delta = 45°$ (open circles). $\Delta = 0°$ corresponds to the ⟨100⟩ direction and $\Delta = -45°$ or 45° to the ⟨110⟩ direction.

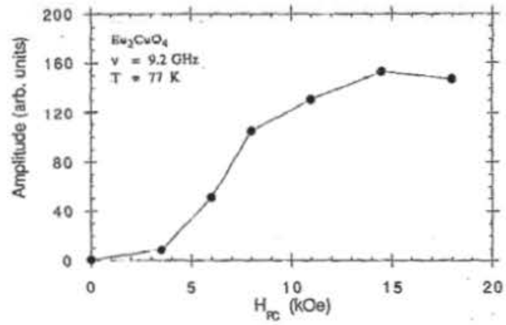

FIG. 3. Maximum microwave absorption signal amplitude as a function of the magnitude of the cooling field H_{FC} applied along a ⟨110⟩ direction.

exhibits a field for resonance, H_r, of ~ 500 Oe and a peak-to-peak linewidth ΔH of ~ 50 Oe. Note that the signal amplitude is a very sensitive function of the angle Δ of H_{dc} within the CuO_2 plane.

This dramatic in-plane anisotropy is further illustrated in Fig. 2, where we present the amplitude of the MAS as a function of Δ for two directions of cooling field, H_{FC}. When the sample is field cooled from above T_N to 77 K along any ⟨110⟩ direction, the MAS is only observed near the perpendicular ⟨110⟩ direction. In Fig. 2 the open circles correspond to cooling in a field H_{FC} along the ⟨110⟩ direction, $\Delta = 45°$. In this case the signal is only observed near the perpendicular ⟨110⟩ direction, $\Delta = -45°$. If the sample is cooled in a field pointing in an arbitrary direction (e.g., $\Delta = -6°$) within the CuO_2 plane, the MAS is then observed in both ⟨110⟩ directions (solid circles in Fig. 2), with an amplitude which is largest for the ⟨110⟩ direction most nearly perpendicular to the direction of H_{FC}.

The MAS amplitude also depends strongly on the strength of the cooling field. This dependence is illustrated in Fig. 3. The sample was cooled in a field H_{FC} applied along a ⟨110⟩ direction and the maximum MAS amplitude was then measured along the perpendicular ⟨110⟩ direction. No signal was observed for $H_{FC} = 0$. From the figure it appears that a cooling field of about 14 kOe is necessary to fully develop the MAS amplitude.

In addition to not observing the MAS if the Eu_2CuO_4 is cooled in zero field, the MAS is also not observed if the microwave magnetic field h_{rf} is either perpendicular to the CuO_2 plane or parallel to the measuring field H_{dc}.

The field for resonance H_r and the linewidth ΔH of the MAS are both found to be independent of the direction or strength of the cooling field. H_r, ΔH, and the signal intensity are also found to exhibit only a weak temperature dependence between 77 and ~ 150 K. However, as the temperature is increased above 150 K, H_r and ΔH both increase, and the signal intensity decreases, until the signal finally disappears at T_N.

H_r and ΔH are found to exhibit an out-of-plane anisotropy identical to the low-field and midfield absorptions studied previously.[1] They both follow a $1/\cos\theta$ dependence where θ is the angle of the applied magnetic field H_{dc} to the CuO_2 plane. This out-of-plane anisotropy, and the nonobservability of the MAS for h_{rf} parallel to H_{dc}, is suggestive that the MAS is due to EPR of the Cu–O system.

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[1] S. B. Oseroff, D. Rao, F. Wright, D. C. Vier, S. Schultz, J. Thompson, Z. Fisk, S.-W. Cheong, M. F. Hundley, and M. Tovar, Phys. Rev. B 41, 1934 (1990).